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RATIO OF CARBON ISOTOPES IN METEORITES

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The ratio of isotopes in cosmic bodies can be used to determine whether they belong to a certain material system in the universe. It is highly improbable that two natural bodies belonging to two different systems of matter-formation will have identical isotopic composition of their elements; conversely, it is highly probable that bodies having identical composition of elements have identical or common evolution of atoms.

It has been proved (2) that there are large accumulations of carbon in our Galaxy which differ sharply in composition from terrestrial carbon. Study of molecular bands in spectra of carbon stars (1-3) has shown that carbon in the atmosphere of these stars is ten times richer in the heavy isotope C13 than carbon in terrestrial objects. Thus, the evolution of nuclear matter of this class of stars followed a path different from that of the evolution of terrestrial matter. It is logical to assume that the isotopic composition of matter in different stars is generally not identical, but varies according to their evolution. Carbon is the first element for which this difference has been experimentally established.

The composition of meteoritic carbon is of special interest for this very reason although any other element is also suitable for the general isotopic characteristics of meteoritic matter.

Results of a comparative study of the isotopic composition of carbon in 39 meteorites are published in this paper. The 39 specimens included 26 stone, 10 iron, and 3 iron-stone meteorites. The iron and olivine

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fractions were analyzed separately in the latter. Samples of  $\text{BaCO}_3$  with carbon separated from the meteorites were prepared for isotopic analysis. To do this, from 0.3 to 3 grams of crushed specimens were weighted out and calcined in an oxygen stream in a quartz tube at 900 degrees centigrade. Carbon dioxide, passing through cupric oxide heated to 500 degrees centigrade, was absorbed by the barite solution. The excess  $\text{Ba}(\text{OH})_2$  was titrated with 0.1 N HCl to determine the total carbon content in the samples. The  $\text{BaCO}_3$  precipitate was filtered, washed, and dried at 110 degrees centigrade. The  $\text{BaCO}_3$  samples thus prepared were used for isotopic analysis with the help of a gas mass-spectrometer previously described (4). The sensitivity of the instrument had been greatly improved by improving the source and increasing the speed of the ions.

To analyze small portions (about 10 milligrams), the  $\text{BaCO}_3$  samples in tin boats were inserted into the special intake system of the mass-spectrometer. The boats were inverted in a vacuum over concentrated  $\text{H}_3\text{PO}_4$  (with the help of a magnet) and the  $\text{CO}_2$  liberated entered the ion tube of the instrument for analysis. The  $\text{H}_3\text{PO}_4$  was preheated with hot water to speed up dissolution of the weighed portions. All determinations were made so that the samples to be analyzed were alternated in the mass-spectrometer with the same control carbon dioxide of known isotopic composition. Thus, each analysis was "squeezed" between two control  $\text{CO}_2$  determinations; all samples were compared with this standard. This method sharply reduced errors in the isotopic ratio with time caused by instrumental drift. This drift reached 3 percent during analysis (80 determinations of the control  $\text{CO}_2$ ).

In the mass-spectral analysis, we compared not the main peaks of the  $\text{CO}_2$  carbon isotopes with mass 44 and 45, but the small peaks of mass 45 and 46 corresponding to molecules  $\text{C}^{13}\text{O}_2^{16}$  and  $\text{C}^{12}\text{O}^{16}\text{O}^{18}$ .

Since all samples of  $\text{BaCO}_3$  were obtained by identical operations and since the same cylinder oxygen of "air" composition was used for combustion of carbon, it can be considered that all  $\text{BaCO}_3$  samples have the same oxygen composition. Therefore, variations in ratio of the two small peaks were caused only by differences in the carbon isotopic ratio  $\text{C}^{12}:\text{C}^{13}$ . The absolute value of the  $\text{C}^{12}:\text{C}^{13}$  ratio is easily calculated from the ratio of the small peaks  $q = h_{45}:h_{46}$ , if the isotopic composition of the oxygen is known. For "normal" oxygen, the  $\text{C}^{12}:\text{C}^{13}$  ratio is  $100/(0.401 q - 0.08)$ .

The control carbon dioxide was also compared according to all three peaks 44, 45, and 46 with the carbon dioxide of limestones (4 samples), which have been shown (7) to be richer in the heavy isotope  $\text{C}^{13}$  than other terrestrial objects. All data cited below is thus referred to the  $\text{C}^{12}:\text{C}^{13}$  ratio of limestones, which equals 89.2.

We also analyzed (according to the small peaks) three samples of oil, which has been shown (7, 8) to have the lowest content of the heavy isotope  $\text{C}^{13}$ , and several other terrestrial objects. The probable relative error of the data cited is  $\pm 0.3$  percent; the error can reach 1 percent only for individual data.

The results obtained are shown in Table 1. The same data, divided into five groups according to the  $\text{C}^{12}:\text{C}^{13}$  ratio, is shown in Figure 1 in the form of a variation curve. The curve shows graphically, but not by individual meteorite, the spread of the isotopic ratio of meteorites.

The average value of the  $\text{C}^{12}:\text{C}^{13}$  ratio is practically equal for stone and iron meteorites. Thus, there is no isotopic distinction between these two phases of matter, with an accuracy to 0.1 percent of the  $\text{C}^{12}:\text{C}^{13}$  ratio. This was confirmed by additional experiments with the three pallasites noted

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by asterisks in Table 1. The isotopic composition of carbon in olivine and in the magnetic iron fraction for all three pallasites was identical with an accuracy down to 0.1 percent, no interphase distinction of carbon isotopes in the olivine-iron contact zone being observed. Apparently, the composition of meteorite carbon in both its main forms of occurrence, i.e., in carbides and in graphite (5), is identical.

The  $C^{12}:C^{13}$  ratio varies from 89.6 to 91.8 for all meteorites. Thus, the extreme spread does not exceed 2.5 percent. About 70 percent of all samples did not deviate by more than 0.5 percent from the average composition; more than 1 percent deviation from the average was observed in only three samples. Taking the probable error of the analyses (0.3 percent) and the Gaussian scattering of the data caused thereby into consideration, we can assume that the real dispersion of the  $C^{12}:C^{13}$  ratio for meteorites is less than the figure given.

No clear correlation was observed between the total content of carbon and its isotopic composition through a wide interval of carbon concentrations (from 0.01 to 2.6 percent). Carbonaceous chondrites (the last six stone specimens in Table 1) are in no way isolated on the monotonic background of isotopic ratios. We note only a tendency for carbon to "become heavy" in low-carbon iron meteorites.

We introduce data obtained from analysis of several terrestrial objects in Table 2 for comparison.

Comparison of Tables 1 and 2 shows that meteoritic carbon in composition is intermediate between carbon of oil and carbon of carbonates. Meteorite carbon is more homogeneous than carbon of the earth's biosphere.

The  $C^{12}:C^{13}$  ratio in natural objects was studied in more detail by Murphey and Nier (7, 8). They also analyzed seven meteorites (mostly iron). The average  $C^{12}:C^{13}$  ratio obtained by these authors for meteorites and oils was 0.5-0.8 percent higher than ours. Their data indicated that meteoritic carbon was intermediate between carbonate carbons and petroleum carbons of the earth and closer in composition to carbon of animal products.

Because of the marked dispersion in the isotopic ratio of carbon in the earth's biosphere shown by available data, the average composition of primary terrestrial carbon can be evaluated only approximately. If we assume (1) that all carbonate and organogenous carbon of the biosphere was derived from primary carbon of the earth's crust and (2) that the mass of carbonate carbon is of the same order or, in any case, not greater (9) than the mass of other organogenous forms of carbon, then the  $C^{12}:C^{13}$  ratio for primary carbon of the earth must be close to its average value for both forms of terrestrial carbon, i.e., close to 90.3 percent. This figure differs from the average meteoritic ratio by only 1/3 percent.

All data on meteoritic isotopy obtained up to the present by various authors and different methods indicates without exception that meteoritic elements do not differ from terrestrial elements in their composition. This result was obtained in the study of ten different elements (4). Only about 35 samples have been subjected to this isotope check by all authors.

Thus, the earth's matter and all 35 meteorites previously studied belong to a single system of formation and have an identical nuclear genesis.

Naturally, this conclusion does not deny the possibility of finding a "star" meteorite with a composition different from that of elements on the earth. But the empirical probability of finding one or, more correctly, the upper limit of this probability  $W$ , is inversely proportional to the number  $n$  of meteorites subjected to such an isotope check. Obviously, the upper limit of this probability is  $W \leq 1/n$ .

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The data cited on carbon isotopy demonstrates the genetic unity of terrestrial and meteoritic matter; it also increases the previous number  $n$  by 35 units, i.e., doubles it in that four of the 39 samples were analyzed previously (4) and consequently, the maximum value of  $W$  is  $1/70$  instead of  $1/74$ .

The actual value of this probability is unknown and may be close to zero. Recent data of meteoric astronomy has also begun to give definite answers as to whether meteorites belong to the solar system (10, 11), although very recently hyperbolic orbits and, consequently, a stellar origin have been ascribed to many meteorites.

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[Tables and figure are appended.]

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Table 1. Content and Composition of Carbon in Meteorite  
Samples Obtained from the Collection of the Academy of Sciences USSR

Meteorites	Total Con- tent (%)	C <sup>12</sup> :C <sup>13</sup>
Stone Meteorites		
Pavlovka	0.017	90.9
Pervomayskiy Village	.020	90.6
Gross Libental'	.022	90.8
Krasnyy Ugol	.025	90.6
Kunashak	.025	91.2
Okans	.026	90.6
Zhovtnevyy Khutor	.031	91.2
Demino	.031	90.9
Savchenskoye	.033	89.6
Kuznetsovo	.063	90.6
Saratov	.068	90.3
Borodino	.070	91.0
Kashin	.076	90.8
Timokhino	.096	90.9
St. Pes'yanoye	.10	89.9
Petropavlovka	.11	90.0
Berdyansk	.12	90.9
B. Korta	.16	90.7
Sevryukovo	.15	90.7
Stavropol'	.17	90.6
Kainsaz	.46	90.6
Groznyaya	.78	90.3
Cold Bokkeveld	1.95	90.6
Nov. Urey	2.13	90.9
Mingey	2.5	90.9
St. Boriskino	2.65	89.9
Average		90.61
Iron Meteorites		
Abakan	0.011	90.9
Sikhote-Alin'	0.17	89.8
Dorofeyevka	.023	90.4
Niro	.026	89.6
Veliko-Nikolayevskiy	.047	90.4
Boguslavka	.048	89.9
Lipovskiy Khutor*	.053	91.5
Il'inskaya Stanitsa	.054	91.2
Bragin*	.056	91.8
Avustinovka	.067	91.2
Pallasovo Zhelezo*	.084	90.6
Chinge	.26	90.6
Chebarkol	.43	90.9
Average		90.68

(\*) Used in additional experiments as noted in text above.

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Table 2. Isotopic Composition of Carbon of Terrestrial Objects

Article	$C^{12}:C^{13}$
Oil, Andizhan	92.4
Oil, Surakhany	91.4
Oil, Sterlitamak	91.2
Cotton	90.6
Beet Sugar	90.9
Limestones (4 samples)	$89.2 \pm 0.2$
Shells (Kola-Bay)	89.0
BaCO <sub>3</sub> Reactive (Standard)	90.9

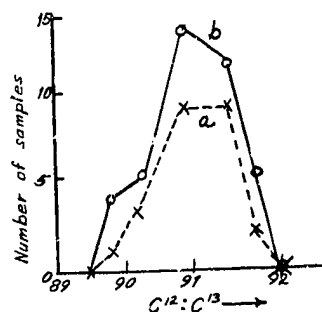


Figure 1. Distribution of Meteorites With Respect to Isotopic Composition:  
 a - Stone, b - all meteorites studied

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